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The exchangeability and leachability of metals from select green roof growth substrates

Sarah Alsup · Stephen Ebbs · William Retzlaff

Abstract Batch extraction and leaching studies were conducted with potential green roof substrates (e.g., Axis, Arklayte, coal bottom ash, Haydite, Lassenite, lava rock, and composted pine bark). The results indicated that these materials would not likely be sources of Cr, Cu, Fe, Ni, or Zn and that Lassenite would be considered a source of Mn if the leachate concentrations were compared to USEPA drinking water standards for these elements. Lassenite would not be a source of Mn if the data was compared to a USEPA standard for Mn toxicity to aquatic life. All of the substrates tested leached Cd and/or Pb concentrations that exceeded the USEPA water quality standards at least once during the 6-month leaching study, so these materials may be potential sources of Cd and Pb in green roof storm water runoff. The leaching of Cu, Cd, Fe, Mn, Pb, and Zn was differentially influenced by time and/or the presence of a single *Sedum hybridum* 'immergrauch' plant. The leaching of Cd, Cu, and Pb displayed complex, three-way interactions between main effects (substrate type and the presence or absence of a plant) and between leaching events. For all substrates except Lassenite, the presence of a *S. hybridum* plant decreased the leaching of Pb over time. The leaching of Cd was generally enhanced by plants for most substrates with time. Collectively the results suggest that changes in the biogeochemical conditions within green roof systems may alter metal solubility, decreasing the leaching of some elements and increasing the leaching of others.

Keywords Green roofs · Heavy metals · *Sedum hybridum* · Vegetated roofs · Water quality

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S. Alsup · S. Ebbs (✉)
Department of Plant Biology, Southern Illinois University Carbondale, 420 Life Science II,
1125 Lincoln Drive, Carbondale, IL 62901-6509, USA
e-mail: sebbs@plant.siu.edu

W. Retzlaff
Department of Biological Sciences, Southern Illinois University Edwardsville,
Box 1651, Edwardsville, IL 62026-1651, USA

Introduction

Green roofs have the potential to relieve urban environmental concerns such as heat island effects, stormwater drainage, and loss of green space. Compared to non-greened roofs, a green roof can provide up to 40% in energy savings and can reduce local ambient temperatures (Niachou et al. 2001). A green roof can also increase storm water retention, up to 40% in some cases (DeNardo et al. 2005). However, these are not the only problems occurring in largely populated areas, cities are also faced with numerous water quality concerns. Anthropogenic pollutants, including pesticides, plant nutrients, and heavy metals present in the urban setting are deposited by rain, picked up by stormwater runoff, and can ultimately enter municipal water supplies and watersheds (Mason et al. 1999). Several studies have demonstrated that conventional roofing materials themselves can be sources of heavy metals that leach pollutants into runoff (Chang et al. 2004; Förster 1996; Gnecco et al. 2005; Göbel et al. 2007; Mason et al. 1999; Schriewer et al. 2008; Zobrist et al. 2000). The capacity of green roof systems to sequester these pollutants, and therefore improve water quality, has been a question of recent interest. Several studies have shown that green roof systems have the capacity to retain pollutants introduced through rainwater (Berndtsson et al. 2009; Berndtsson et al. 2006; Köhler et al. 2002; Steusloff 1998), but the extent of retention is dependent upon factors such as the water retention capacity of the roof system (Berndtsson et al. 2006; Steusloff 1998) and whether the roof was designed with pollutant retention in mind (Berndtsson et al. 2009).

Whether green roofs will act as sources or sinks for pollutants will depend primarily upon the nature of the substrate in the roof system. Mass, color, and thermal properties are characteristics routinely considered when selecting a substrate as is the capacity of the substrate to serve as a plant growth media. With the increased interest in green roofs, a wide variety of substances are being considered as potential substrates. Manufacturers are exploring the potential use of recycled waste materials for use in green roof systems. As these different substrates are being developed, a question that will have to be addressed is whether these substrates, many of which are derived from highly heterogeneous materials and are largely uncharacterized, will serve as sources for anthropogenic pollutants. However, even if these materials are initial sources of pollutants, this may not necessarily be problematic as it may be possible to readily leach undesirable components from the substrate prior to their use in a green roof system. In addition, the plants in the green roof system may sequester, phytoextract, or phytostabilize some fraction of the pollutants that may be released from the substrate, thereby reducing the concentration of the pollutant leached from the system.

The objectives of the research here were three-fold. The first objective was to characterize the content and exchangability of metals and micronutrients in several substrates being considered for inclusion in green roof systems. Those substrates were coal bottom ash, Axis, lava rock, Lassenite, Haydite, and Arkalyte. Arkalyte and Haydite are substrates that are currently being used in green roof systems (Beatie and Berghage 2004; Retzlaff et al. 2008). Lava rock is a lightweight natural stone material that is available commercially and has been used previously in a soil mixture for a vegetated roof (Emilsson and Rolf 2005). Axis, Lassenite, and bottom ash are materials that have been donated by vendors who are interested in determining if these materials would be suitable substrates for green roof systems. There is little to no published literature characterizing these substrates so the study here represents one of the first efforts to do so. Also included in this experiment was pine bark, which is a readily available organic amendment added to some green roof systems as a soil conditioner to increase the organic matter content and water holding capacity of the substrate (Retzlaff et al. 2008).

The second objective of this study was to examine the potential leachability of elements from these substrates to begin evaluating whether these materials might be sources of pollutants in green roof systems. The leaching experiment simultaneously addressed the third objective, which considered whether the presence of a plant, in this case *Sedum hybridum* ‘immergrauch’, a species belonging to the same genus as several other species which are reported to be facultative CAM plants (Müller et al. 2006) and currently being used in some green roof systems (Forrester 2007; Monterusso et al. 2005; van Woert et al. 2005; Villarreal and Bengtsson 2004), altered the leaching characteristics of these elements from the substrates. The overarching goal of this study was to provide results that would help guide the development and construction of green roof systems to maximize the benefits for the urban environment.

Methodology

Green roof substrates

Most of the substrates used in this study are derived from potentially heterogeneous sources or are proprietary, so there is only a modest amount of information available (e.g., from MSDS sheets or company websites) regarding composition and physical characteristics. The Axis substrate used in this study was commercially available calcined diatomaceous earth containing <1% crystalline silica (cristobalite) and <1% crystalline silica (quartz) prepared by kiln-firing at ~1,800°C, resulting in a porosity of 82%, pore sizes ranging from 0.1 to 1.0 μm and a pH of 7. The commercially available Lassenite used consisted of 70% SiO_2, 10% aluminum silicate (kaolin), and 2% crystobilite. This substrate has 55.4% total porosity, 41.0% capillary porosity and 14.4% air-filled porosity with a pH of 4.65. Haydite is an expanded shale comprised of 59.8% SiO_2, 22.6% Fe_2O_3, and 10.2% CaO with a pH of 7.5. Porosity data for Haydite was unavailable. The Arkalyte was an expanded clay substrate with a proprietary composition. The red lava rock employed in this study was a natural stone quarried from northern New Mexico’s Twin Mountain quarry. This rock was not washed prior to use and may have retained dust and powder from the quarry site within its pores (T. Tharp, personal communication). Unlike fly ash, bottom ash is the heavier material left at the bottom of the furnace after combustion. The properties of bottom ash, including the concentration of heavy metals and micronutrients, vary depending upon the type of furnace, the temperature of combustion, and the initial elemental profile of the coal (Brunner and Monch 1986; Chang and Wey 2006). The coal that produced the bottom ash for this study was a sub-light bituminous coal acquired from the Powder River Basin in Wyoming (M. Bryant, personal communication). While the material used here was not specifically characterized, a typical bottom ash has 70–85% aluminosilicate glass (contains Al, Si, Fe, Ca, Mg, Ti, K), 0–15% crystalline silica, 2–12% mineral Fe, and 0–20% calcium oxides (Chang and Wey 2006). Mean concentrations for elements of interest are 15 mg Cd kg DW^{-1}, 375 mg Cr kg DW^{-1}, 2,818 mg Cu kg DW^{-1}, 45,874 mg Fe kg DW^{-1}, 1,288 mg Pb kg DW^{-1}, and 4,229 Zn mg kg^{-1} (Jung et al. 2004).

Total acid extractable and exchangeable metals in the substrates

Approximately 50 g of each of the six substrates (Lassenite, Haydite, Arkalyte, bottom ash, Axis and lava rack) and one amendment (composted pine bark) were ground and passed through a 2 mm sieve. Composted pine bark was included here because it is used as a soil conditioner for some green roof systems and this substrate’s potential contribution to metal leaching and retention was also of interest. Approximately 1 g of each substrate was subjected to total acid extraction according to EPA method 3050b (http://www.epa.gov/epaoswer/hazwaste/test/pdfs/3050b.pdf), using nitric acid (trace metal grade) and hydrogen peroxide. The acid extracts were recovered, centrifuged or filtered as necessary to remove residual particulates, and diluted to 50 mL with deionized water. Exchangeable elements were estimated by mixing each substrate (1:10 w/v) with of 0.5 N ammonium acetate + 0.02 M EDTA (pH 4.65) and shaking the slurry for 16 h. The extract was recovered by sequential vacuum filtration through Whatman No. 1 and No 42 filter paper with the final filtrate passed through 0.45 μm cellulose filters. For both of these procedures, four repetitions of each substrate were included.

Leaching of metals from planted and non-planted green roof substrates

The leaching study used a completely randomized design consisting of seven substrates, a planting treatment (i.e., plants present or absent), and eight replications (112 pots). The substrates used were Lassenite, Haydite, Arkalyte, bottom ash, lava rock, Axis, and a 4:1 blend by mass of bottom ash and Axis. Each substrate was used directly as provided by the manufacturer with no modification of particle size and no wetting prior to this experiment. Each substrate was amended with composted pine bark at 3:1 ratio of substrate to pine bark. The final mass in the pots for each type of substrate is shown in Table 1. The square pots used were filled with the indicated mass of each substrate and then fertilized with 1.41 g of a blend of three fertilizers, which was a 1:1:1 (v/v/v) mixture of IBDU (1,1[1] isobutylidene-diurea), Osmocote®, and Nutricote® to achieve a final N-P-K ratio of 15-9-12. Fertilizers are a critical component to provide essential nutrients for plant survival, growth, and sustainability of green roof systems. The rate and frequency of fertilizer application is important, both for plant survival and performance of the green roof, including *Sedum* spp. roofs (Emilsson et al. 2007; Rowe et al. 2006).

Half of the pots were planted with one *Sedum hybridum* 'immergrauch' plug placed in the center of the pot. These plants were propagated from cuttings taken from stock plants from the greenhouses at Jost Greenhouses (St. Louis, MO) throughout the months of June to September (V. Jost, personal communication). Cuttings were placed into a porous Peat-Lite® media and subjected to a light mist three times each day. After the softwood cuttings had taken root, the mist was then reduced to harden the plants, and the rooted plants (plugs) were placed into fresh Peat-Lite® soil. The rooted plants were obtained from Jost Greenhouses and transplanted to the center of each green roof substrate pot. The pots were given sufficient water to wet the substrate so the plants could become established, but not

Table 1 The range of particle sizes for each substrate used in the leaching experiment, the mean mass of substrate present in the planted and unplanted pots of each substrate type (+/− standard error, $n=16$), and the volume of water used to saturate each substrate prior to each leaching event. All pots were leached with 0.25 L of deionized water

Substrate	Range of substrate particle size (cm)	Mean mass of substrate in pots (g)	Volume of water to saturate substrate (L)
Arkalyte	0.6–1.6	440.9 (5.0)	0.15
Axis	0.2–0.5	303.8 (1.8)	0.30
Bottom ash	0.4–1.8	549.5 (7.6)	0.10
Axis+bottom ash	0.2–1.8	556.1 (4.3)	0.20
Haydite	0.3–2.0	626.4 (5.2)	0.15
Lassenite	0.5–1.5	609.9 (10.1)	0.15
Lava rock	1.3–1.9	513.2 (2.7)	0.15

enough to leach water from the pots. The pots were placed in a phytotron under ambient light supplemented with sodium halide lamps on a 16 h photoperiod. Ambient temperature ranged from 20–25ºC during the course of the experiment. As potentially facultative CAM plants with a high resistance to water stress, the plants received minimal watering once a week during the course of the experiment (6 months) to prevent premature loss of leachate.

The pots were subjected to three episodes of leaching at 2 month intervals. To obtain leachate, each pot was watered to saturation 2 days prior to collection. To insure uniform wetting of each substrate, saturation conditions were imposed by adding distilled water in 50 mL increments to containers placed below each pot and allowing capillary action to draw the water into the pot until water had reached the substrate surface. The total volume of water added is shown in Table 1. These same volumes were used for each of the three leaching events. After these wetting treatments were imposed, the pots were left to stand for approximately 2 days. To collect leachate, 0.25 L of distilled water was added to the surface of each pot to displace the water in the pots. The leachate from each pot was collected and subjected to vacuum filtration with a final filtration through 0.45 μm cellulose filter paper.

Elemental and statistical analysis

Elemental analysis of the various extracts and leachates for Cd, Cr, Cu, Fe, Mn, Ni, Pb, and Zn was conducted using a SpectrAA 220FS Atomic Absorbance Spectrometer (Varian, Palo Alto, CA). The results for each metal from the experiments for total acid extractable or exchangeable metals were subjected to a one-way ANOVA as a function of substrate type. For the leaching experiment, the data were analyzed using a two-way ANOVA with repeated measures. Fixed effects were substrate type and planting status (meaning planted or unplanted) and the repeated measure was leaching event. Data were analyzed using SAS, Version 9.1.3, (SAS Institute 2007) with the PROC GLM procedure.

Results

Total acid extractable and plant exchangeable metals

With the exception of total extractable Cd, there were significant differences in total acid extractable metals between the substrates tested. However, the patterns differed greatly between metals. Total extractable Cd was not significantly different between substrates, ranging from ~0.6 to ~1.6 mg kg DW^{-1} (Fig. 1A). In contrast, there was a 10-fold variation in Cr concentration, ranging from lowest value for Axis to nearly 35 mg kg DW^{-1} for Lassenite. Lassenite and Arkalyte collectively had significantly greater ($p \leq 0.0001$) total acid extractable Cr than the other substrates (Fig. 1B). Pine bark had the highest total acid extractable Cu and Fe concentrations (Fig. 1C and D), values which were significantly higher than the remaining substrates ($p \leq 0.0001$ for both). The Cu and Fe concentrations for the remaining substrates were generally not significantly different, with the exception of Axis for Cu and Lassenite for Fe. The widest range of concentrations was observed for Mn (Fig. 1E). Axis and bottom ash showed total acid extractable concentrations of 10.4 and 16.3 mg kg DW^{-1} respectively while composted pine bark had a mean >300 mg kg DW^{-1} and Arkalyte a mean of >400 mg kg DW^{-1}. With respect to Ni content (Fig. 1F), lava rock had the highest mean concentration (41.4 mg kg DW^{-1}), a value significantly greater than the remaining substrates ($p \leq 0.0001$). Axis was the substrate that showed the lowest acid

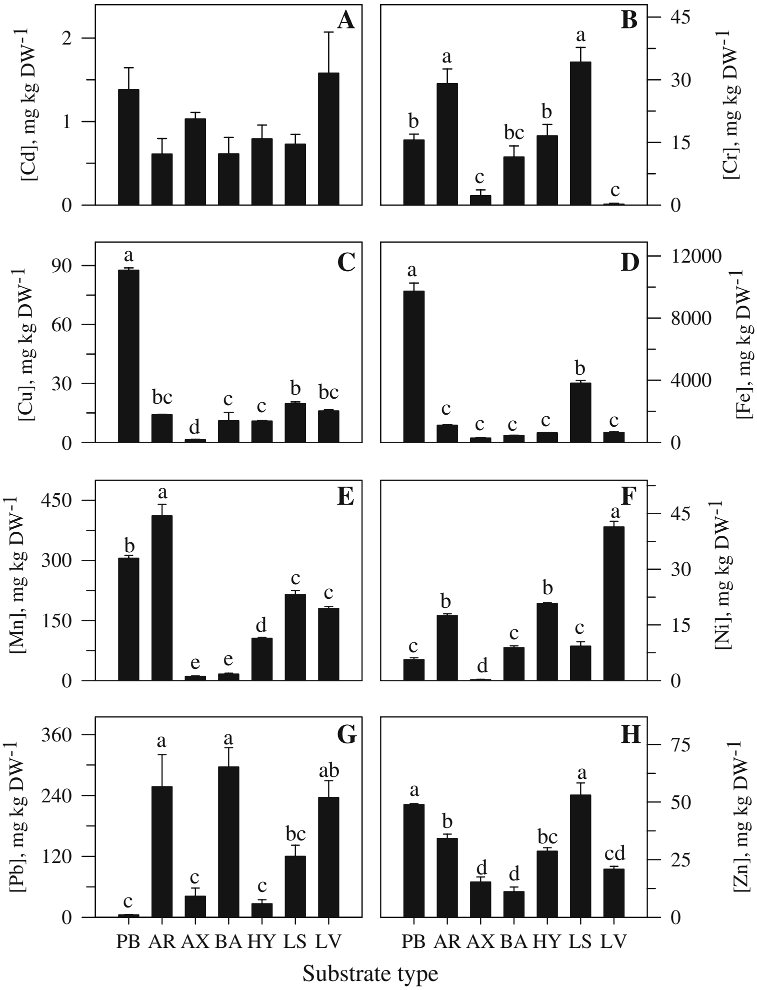


Fig. 1 Total acid extractable Cd (**A**), Cr (**B**), Cu (**C**), Fe (**D**), Mn (**E**), Ni (**F**), Pb (**G**), and Zn (**H**) content of the pine bark amendment and the six potential green roof substrates. Data represent the mean and standard error (*n*=4). Within a panel, different letters are used to denote significant differences between substrates. There was no significant difference in total acid extractable Cd between substrates. PB=pine bark; AR=Arkalyte; AX=Axis; BA=bottom ash; HY=Haydite; LS=Lassenite; LV=lava rock

extractable Ni concentration. Arkalyte, bottom ash, and lava rock showed the highest acid extractable Pb concentrations (>200 mg kg^{-1}, Fig. 1G), followed by Lassenite with a mean concentration of 53.1 mg kg DW^{-1}. The acid extractable Pb from the remaining substrates was <50 mg kg DW^{-1}. Composted pine bark and Lassenite had the highest Zn concentrations (~50 mg kg DW^{-1}), ranging down to values of ~15 mg kg DW^{-1} for Axis and bottom ash (Fig. 1H).

Of these eight elements examined, all fell within the range of total concentrations typically encountered for surface soils. For example, the Cu and Ni concentrations of surface soils generally range from 1–100 mg kg DW^{-1} (Kabata-Pendias 2001). The total acid extractable Cu (Fig. 1C) and Ni (Fig. 1F) concentrations observed for the various green roof substrates were at the lower end of this range. Pine bark was closer to the upper end of the range for Cu concentration. A typical range for soil Cd concentrations is 0.1–2.5 mg kg DW^{-1}, which corresponds to the total acid extractable Cd concentrations observed for all the substrates tested here (Fig. 1A). The Pb concentration for most natural soils is <100 mg kg DW^{-1}, but there are paddy soils, Rendzinas and histosols with naturally-occurring Pb concentrations of up to 280 mg kg DW^{-1} (Kabata-Pendias 2001). Arkalyte, bottom ash, Lassenite, and lava rock were at the upper end, but still within, this range (Fig. 1G).

Exchangeable concentrations of these elements were all considerably lower than the corresponding total acid extractable values. For all elements except Ni and Pb, pine bark was the substrate that had the greatest exchangeable concentration (Fig. 2). For example, plant exchangeable Cd from pine bark (0.17 mg kg DW^{-1}, Fig. 2A) was significantly

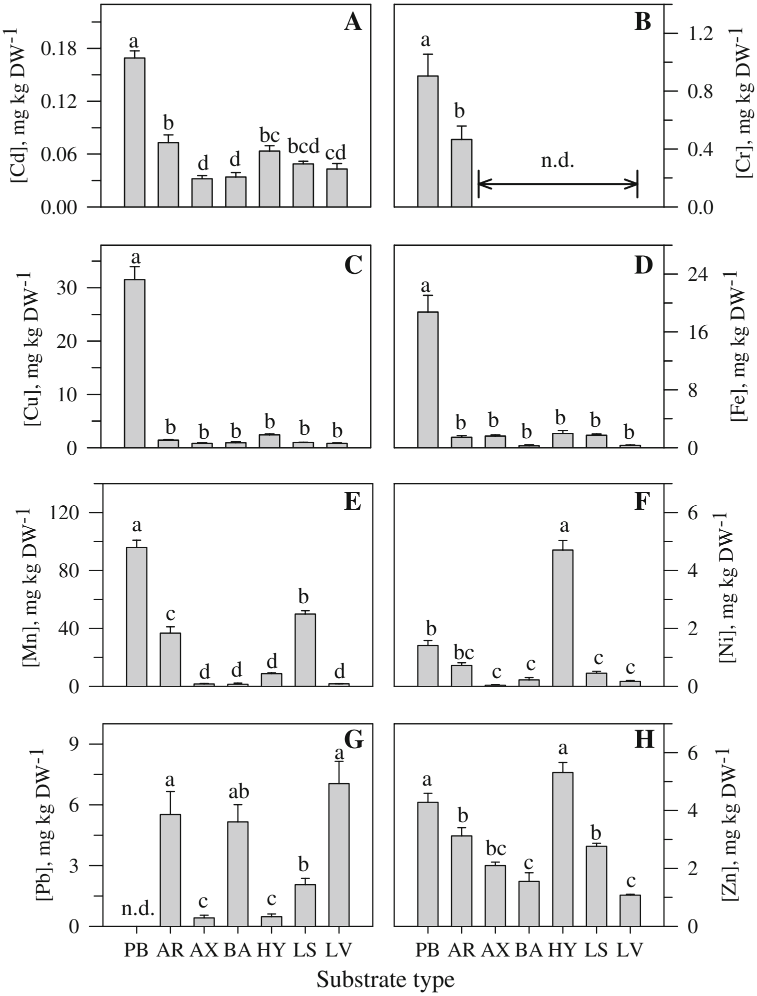


Fig. 2 Exchangeable Cd (**A**), Cr (**B**), Cu (**C**), Fe (**D**), Mn (**E**), Ni (**F**), Pb (**G**), and Zn (**H**) from the pine bark amendment and the six potential green roof substrates. Data represent the mean and standard error (*n*=4). Within a panel, different letters are used to denote significant differences between substrates. Substrate codes are shown in the legend for Fig. 1 (n.d.=none detected)

greater than all other substrates ($p \leq 0.0001$) while the remaining substrates had values 3-fold lower. Similar patterns were observed for Cu (Fig. 2C), Fe (Fig. 2D), and Mn (Fig. 2E), although the magnitude of the difference between the value for pine bark and the remaining values was not the same across these elements. For Cr, pine bark and Arkalyte were the only substrates that had Cr concentrations above the limit of detection (0.90 mg kg^{-1}and 0.47 mg kg^{-1}, respectively, Fig. 2B). Haydite had the highest concentration of exchangeable Ni (4.71 mg kg^{-1}, Fig. 2F), but pine bark had the second highest concentration. Arkalyte, bottom ash, and lava rock had the highest mean concentrations of Pb (5.5–7.1 mg kg DW^{-1}). Exchangeable Zn was highest in Haydite and composted pine bark (5.3 and 3 mg kg DW^{-1}, respectively), values which were no more than 2 to 3-fold higher than those observed for the remaining substrates.

When the total acid extractable and exchangeable concentrations for each metal were compared, the data demonstrated that the plant exchangeable concentrations were consistently no more than 10–15% of the total acid extractable concentration. In numerous cases, such as for Cr and Fe, the exchangeable concentration of an element was negligible as compared to the total acid extractable concentrations. The exceptions to these trends were Cu from pine bark (31.5 mg kg DW^{-1}), Mn from pine bark and Lassenite (95.9 mg kg DW^{-1} and 50.0 mg kg DW^{-1}), and Ni from Haydite (7 mg kg DW^{-1}), which represented 36, 31, and 22% of the total acid extractable concentration, respectively.

Metal leaching from green roof substrates

The leaching behavior of each substrate in terms of the volume of water displaced varied significantly as a function of substrate type, planting status, and leaching event ($p \leq 0.002$), but there were no significant interactions between the variables. While significant, the magnitude of the differences was only slight in each case. Across all three leaching events, Arkalyte, bottom ash, Axis + bottom ash, and Haydite discharged the largest volume of leachate (~0.16 L), followed closely by lava rock (~0.15 L), and ~0.13 L leached from the remaining two substrates (Lassenite and Axis, Fig. 3A). The presence of a plant in this experiment significantly reduced the amount leached across all pots, but only by 0.01 L (Fig. 3B). The most noticeable trend was that there was a significant increase in the amount leached across all substrates as a function of leaching event (Fig. 3C). The pots, regardless of the presence of a plant, leached ~0.12 L during the first event and increased to 0.19 L by the third leaching event. Since the same volumes of water were used to saturate each pot (Table 1) and then leach the pot (0.25 L) for each event, and all pots were watered with approximately the same amount of water during the course of the experiment, this may indicate a change in the substrates over time. However, given the differences in the nature of each substrate and the lack of a significant interaction between time and substrate, this is not likely. The only common factor across the substrate types was the presence of pine bark, so this may have been the material that contributed to the difference in leaching with time observed during the experiment.

No Cr was detected in the leachates from any pots in this study (data not shown). Iron showed two-way interactions for each pair of main effects ($p \leq 0.006$, Fig. 4) with the pots containing the Axis + bottom ash mixture driving each interaction. For example, the Fe concentration in the second leaching from these pots was significantly higher than all other treatments (Fig. 4A). The planted pots containing this substrate mixture leached significantly more Fe than the other treatments for the other two-way interactions (Fig. 4B, C). For Mn, Ni, and Zn, the only significant differences observed were for

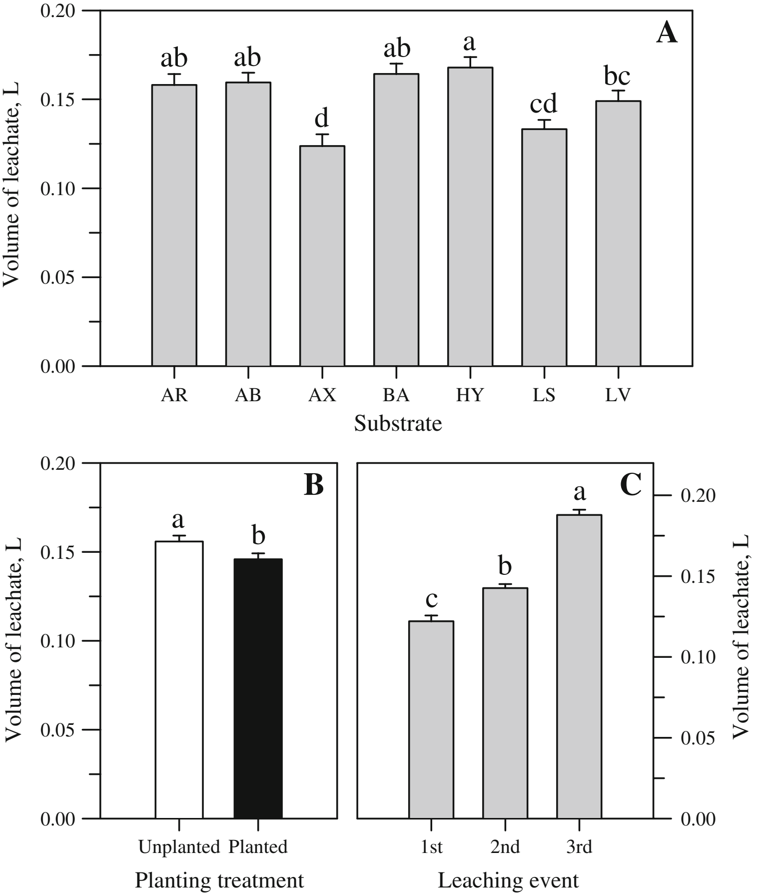


Fig. 3 The volume of leachate from the pots of green roof substrate (substrate+composted pine bark, 3:1 ratio) as a function of substrate type (**A**), whether the pots were planted with a single *Sedum hybridum* 'immergrauch' plant or not (**B**), or leaching event (**C**). Data represent the mean and standard error, with $n=48$ for (**A**), $n=168$ for (**B**), and $n=112$ for **C**. Within a panel, different letters are used to denote significant differences between substrates. AR=Arkalyte; AB=Axis+bottom ash; AX=Axis; BA=bottom ash; HY=Haydite; LS=Lassenite; LV=lava rock

single main effects. There were significant differences in the concentration of all three elements in the leachate (Fig. 5). Lassenite leached significantly more Mn than all other substrates ($p \leq 0.0001$, Fig. 5A). The magnitude of the difference was substantial, with a Mn concentration >350 μg L^{-1} for the Lassenite pots and values of <45 μg L^{-1} for all other substrates. The leaching of Ni from Arkalyte and Haydite pots was 5–10-fold greater than all other substrates ($p \leq 0.0001$, Fig. 5B). There were also significant differences in Zn leaching between substrates, but the range across substrates was much less than that observed for Mn and Ni ($p \leq 0.0001$, Fig. 5C). Significant differences were observed for the leaching of Mn and Zn from pots as a function of leaching event. The third leaching event produced the highest concentration of Mn, with the second leaching event producing the lowest concentration ($p \leq 0.04$, Fig. 6A). Leaching of Zn showed a first flush (Mason et al. 1999) pattern with the highest concentration obtained from the first leaching and lower concentrations for the remaining two ($p \leq 0.0001$, Fig. 6B).

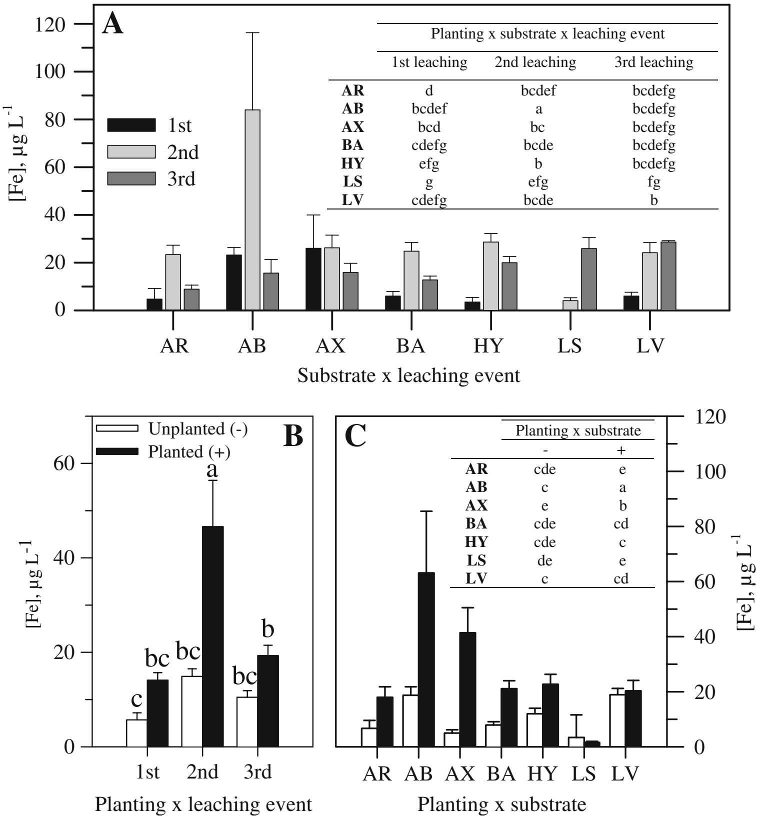


Fig. 4 Leaching of Fe from the pots of green roof substrates (substrate+composted pine bark, 3:1 ratio) as a function of the two-way interactions between substrate and leaching event (**A**), leaching event and the presence or absence of a plant (**B**) or the interaction between substrate type and the presence or absence of a plant (**C**). Data represent the mean and standard error, with $n=16$ for (**A**), $n=56$ for (**B**), and $n=24$ for (**C**). Within (**B**), different letters are used to denote significant differences between substrates. For (**A**) and (**C**), the post hoc lettering for each two-way interaction is shown in an inset table. Substrate codes are shown in the legend for Fig. 3

The data for Cd, Cu, and Pb were the most complex as there were three-way interactions between the main effects for each element. The concentrations of Cd in the leachate were highly variable yet significantly different ($p \leq 0.0001$) across the three leachings for the substrates, but with few consistent trends (Fig. 7). Planted Lassenite and lava rock pots showed a consistent leaching of Cd across the three leaching events, a pattern also observed for the unplanted Axis pots. Unplanted Lassenite, lava rock, and Arkalyte showed a first flush of Cd, and then significantly lower Cd concentrations subsequently. Perhaps the most obvious trend was that by the third leaching, all of the planted pots were leaching Cd while the unplanted pots, with the exception of Axis and the Axis + bottom ash mixture, showed less metal in the leachate than was obtained during the previous leaching events. The highest concentrations of Cd observed were from the Axis and Lassenite substrates (>6 $\mu g\ L^{-1}$). Most of the remaining pots had Cd concentrations of ~2.5 $\mu g\ L^{-1}$ or had concentrations <0.5 $\mu g\ L^{-1}$. For the significant differences for leachate Cu ($p \leq 0.03$, Fig. 8), concentrations ranged from <0.2

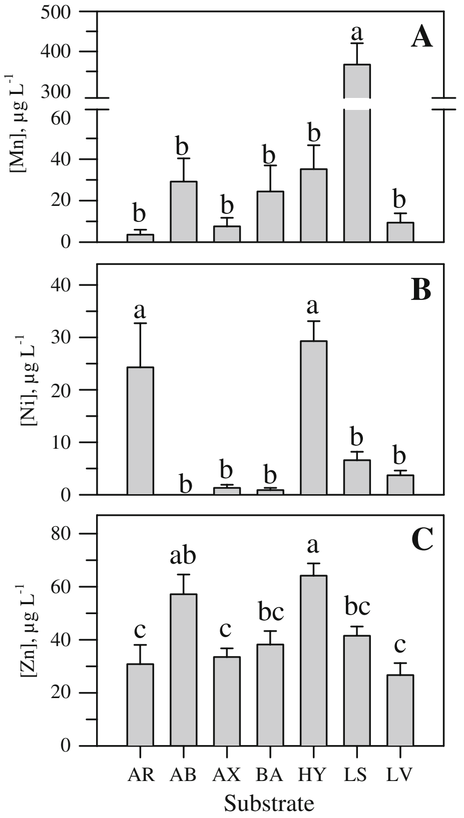


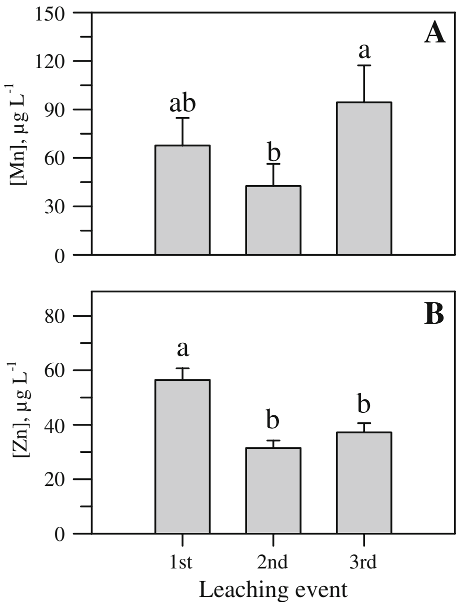
Fig. 5 Leaching of Mn (**A**), Ni (**B**), or Zn (**C**) from the pots of green roof substrates (substrate+composted pine bark, 3:1 ratio) as a function of substrate type. Data represent the mean and standard error, with $n=48$ for each panel. Within a panel, different letters are used to denote significant differences between substrates. Substrate codes are shown in the legend for Fig. 3

to >15 μg L^{-1}. There was a first flush of Cu followed by a general decrease in leachate concentration for the latter two leaching events, with Haydite and lava rock as two exceptions. The concentration of Cu in the leachate from Haydite and lava rock pots did not differ significantly across the three leaching events and, like the other substrates, showed no clear trend with respect to the presence or absence of a plant.

The significant three-way interaction ($p \leq 0.0001$) between main effects produced some interesting results for Pb (Fig. 9). During the first leaching event, Pb was found in the leachate from the planted pots of bottom ash, Axis, and both the planted and unplanted Axis+bottom ash mixture. For the subsequent two leachings, little to no Pb leached from these pots. For the other substrates, leaching was enhanced initially by the presence of a plant, but subsequently there was negligible Pb if a plant was present. The unplanted pots of Arkalyte, Axis, Axis+bottom ash, Haydite, showed the opposite trend. In this case, Pb in the leachate went from <5 μg L^{-1} during the first leaching to >150 μg L^{-1} for these pots after the second or third leaching. Little Pb leached from the Lassenite and lava rock pots during the first leaching, but there was a significant increase in Pb in the leachate during the second and third leachings. For the lava rock, the presence of a plant either had no effect or decreased Pb leaching, while for Lassenite the presence of a plant significantly increased the release of Pb.

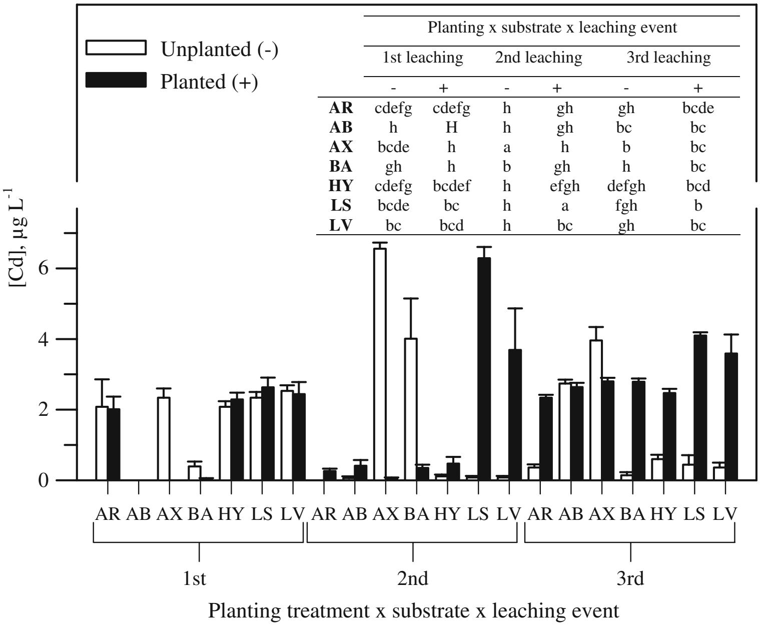
The maximum concentrations of Cu, Fe, Mn, Ni, and Zn observed in the leachate from all of the substrates used here (Table 2) were similar to those that have been

Fig. 6 Leaching of Mn (**A**) or Zn (**B**) from the pots of green roof substrates (substrate+composted pine bark, 3:1 ratio) as a function of leaching event. Data represent the mean and standard error (n=112). Within a panel, different letters are used to denote significant differences between substrates. Substrate codes are shown in the legend for Fig. 3



obtained from studies of runoff from conventional roofing materials (Chang et al. 2004; Gnecco et al. 2005; Göbel et al. 2007; Mason et al. 1999; Zobrist et al. 2000) and from some vegetated roofs (Berndtsson et al. 2009; Berndtsson et al. 2006; Göbel et al. 2007). The values obtained here for Cu and Fe were similar to those from other studies of runoff from convention roofs and did not exceed the USEPA Recommended Water Quality Criteria for that element (USEPA 1999). When the maximum leachate Cu concentration was compared to the USEPA standard,≤6% of the samples collected exceeded that value. Leachate from two types of shingled roofs as well as aluminum and galvanized iron roofs exceeded the same criteria 60–78% of the time (Chang et al. 2004). The other roof types indicated in Table 2, including the vegetated roofs, all had mean Cu concentrations that exceeded the USEPA criteria. A similar pattern was observed for Zn. Only 4–5% of leachate samples here exceeded the USEPA standard, while a considerably higher fraction of samples from both conventional and vegetated roofs apparently exceed that value (Chang et al. 2004). For example, the same roof types noted above for Cu exceeded the Zn standard 99.5 to 100% of the time. Although the Ni concentrations in the leachate were >20-fold higher than reported for other roof types, the maximum value observed in the leachate was still 4-fold below the USEPA standard. The range of Mn concentrations observed in the leachate here were quite variable, spanning from <0.01 to 1,734 µg L^{-1}. While most other studies with conventional or vegetated roofs have values that do not or marginally exceed the USEPA standard, the values here exceeded that standard 16–24% of the time for planted and unplanted pots, respectively, but predominantly from the Lassenite substrate.

The maximum leachate concentrations of both Cd and Pb exceeded the mean values in the literature for roof runoff (Table 2). With respect to Pb, reported mean values from conventional roofs do not exceed 100 µg L^{-1}, although a maximum value of 700 µg L^{-1} was reported for a wood shingle roof receiving rainfall with a mean Pb concentration of 30 µg L^{-1} (Chang et al. 2004). Aside from this maximum concentration, the values here for Pb are up to 100-fold higher that what has been observed previously from conventional and



	Planting x substrate x leaching event					
	1st leaching		2nd leaching		3rd leaching	
	-	+	-	+	-	+
AR	cdefg	cdefg	h	gh	gh	bcde
AB	h	H	h	gh	bc	bc
AX	bcde	h	a	h	b	bc
BA	gh	h	b	gh	h	bc
HY	cdefg	bcdef	h	efgh	defgh	bcd
LS	bcde	bc	h	a	fgh	b
LV	bc	bcd	h	bc	gh	bc

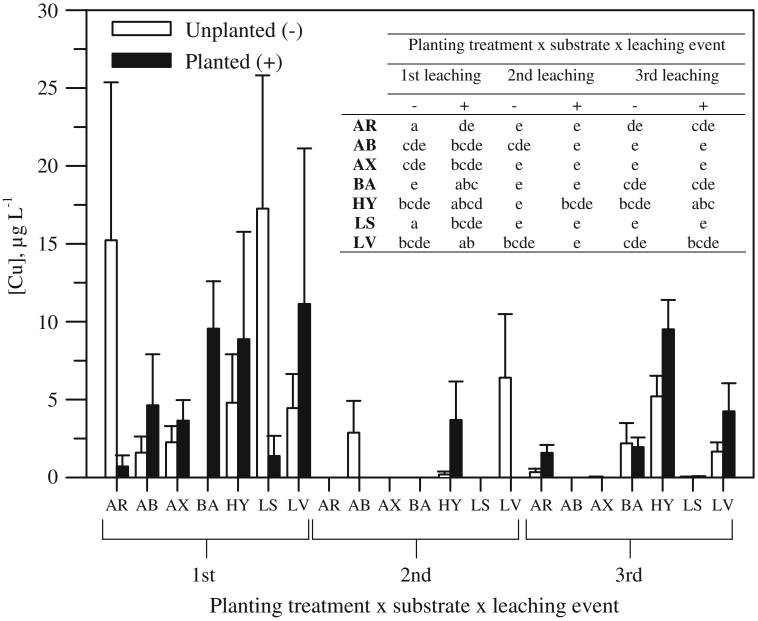
Fig. 7 Leaching from Cd from the pots of green roof substrates (substrate+composted pine bark, 3:1 ratio) as a result of the three-way interaction between substrate type, leaching event, and the presence or absence of a single *Sedum hybridum* 'immergrauch' plant. Data represent the mean and standard error (n=8). The different letters in the inset table are used to denote significant differences between all treatment combinations. In instances where no Cd was detected in the leachate, no data bars are shown for that particular combination of treatment variables. Substrate codes are shown in the legend for Fig. 3

green roofs. The percentage of pots with leachate exceeding the USEPA standard was 45.4% across all substrates and treatments. The presence of a plant restricted Pb leaching as only 29.3% of pots with plants produced leachate Pb concentrations in excess of the USEPA standard while the number was double for the unplanted pots (60.3%). The maximum Cd concentration observed in the leachate was ~4-fold higher than mean values reported in the literature (Table 2). Across all substrates and treatments, ~40% of samples exceeded the USEPA standard of 2 µg L^{-1}. Contrary to the data for Ni and Pb, pots with plants produced leachate that exceeded the USEPA standard for Cd more often than the unplanted pots (49.1 and 30.4%, respectively).

Discussion

Acid extractable and exchangeable elements associated with green roof substrates

The total acid extractable concentrations varied significantly between substrates for all elements examined except Cd (Fig. 1) yet overall the concentrations were not different from those that would be encountered in typical soils (Kabata-Pendias 2001). When considered from that perspective, the use of these substrates in green roofs would ostensibly be no different than utilizing soil. The one element that stood out from these data as a possible concern was Pb. The concentration of Pb in Arkalyte, bottom ash, Lassenite, and lava rock was on the upper end of the range observed for soils. While the batch studies with the finely

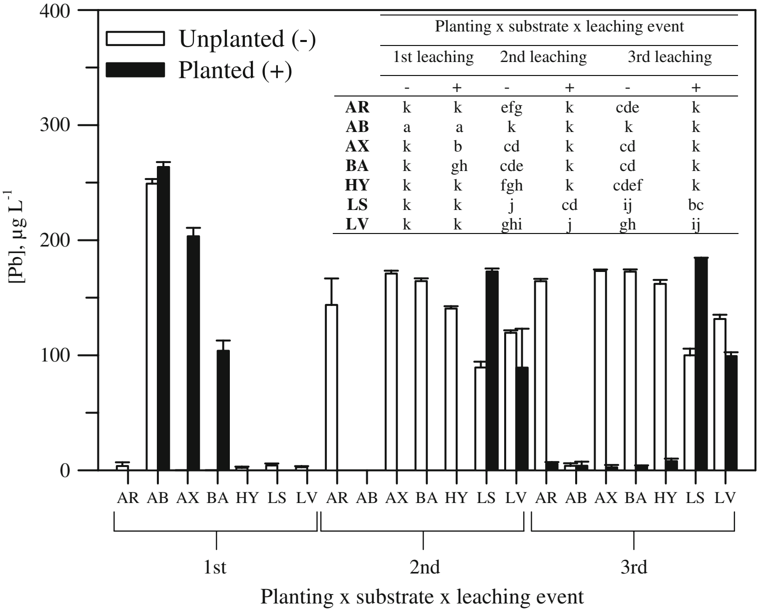


Planting treatment x substrate x leaching event						
	1st leaching		2nd leaching		3rd leaching	
	-	+	-	+	-	+
AR	a	de	e	e	de	cde
AB	cde	bcde	cde	e	e	e
AX	cde	bcde	e	e	e	e
BA	e	abc	e	e	cde	cde
HY	bcde	abcd	e	bcde	bcde	abc
LS	a	bcde	e	e	e	e
LV	bcde	ab	bcde	e	cde	bcde

Fig. 8 Leaching from Cu from the pots of green roof substrates (substrate+composted pine bark, 3:1 ratio) as a result of the three-way interaction between substrate type, leaching event, and the presence or absence of a single *Sedum hybridum* 'immergrauch' plant. Data represent the mean and standard error (n=8). The different letters in the inset table are used to denote significant differences between all treatment combinations. In instances where no Cu was detected in the leachate, no data bars are shown for that particular combination of treatment variables. Substrate codes are shown in the legend for Fig. 3

ground substrates showed that across the eight elements examined <15% of the total elemental content was in a readily exchangeable pool (Fig. 2), the substrates with the higher Pb content were also the substrates that displayed the highest exchangeable Pb. These substrates could therefore be potential sources of Pb in green roof systems if Pb also leaches from the intact, unprocessed materials.

The results for pine bark, which can be used as a soil conditioner in green roof systems (Retzlaff et al. 2008), were curious. Pine bark was the substrate that had the highest concentrations of Cu, Fe, and Zn and the second highest concentrations of Cd and Mn (Fig. 1). As an organic material, pine bark was not expected to have concentrations of these elements higher than the mineral substrates being tested. The pine bark used here originated from a mill in northern Alabama which processes pine trees for the paper and lumber industries (T. Tharp, personal communication). The bark is a surplus material that is held onsite in piles. Bark piles may be present for more than a year before the material is processed onsite by additional parties (e.g., landscaping companies) for future commercial use. During the period of storage on site, the pine bark is unprotected and exposed to the elements. In the absence of additional information, the most logical explanation for the elevated concentrations of these elements is that they were introduced by rain and particulate deposition emanating from natural and industrial activities in the immediate region. The fact that a significant fraction of these elements were exchangeable (Fig. 2) is consistent with adsorption of these elements to the bark surface rather than incorporation into the biomass. These results indicate that organic amendments used in green roof



	Planting x substrate x leaching event					
	1st leaching		2nd leaching		3rd leaching	
	-	+	-	+	-	+
AR	k	k	efg	k	cde	k
AB	a	a	k	k	k	k
AX	k	b	cd	k	cd	k
BA	k	gh	cde	k	cd	k
HY	k	k	fgh	k	cdef	k
LS	k	k	j	cd	ij	bc
LV	k	k	ghi	j	gh	ij

Fig. 9 Leaching from Pb from the pots of green roof substrates (substrate+composted pine bark, 3:1 ratio) as a result of the three-way interaction between substrate type, leaching event, and the presence or absence of a single *Sedum hybridum* 'immergrauch' plant. Data represent the mean and standard error ($n=8$), and different letters in the inset table are used to denote significant differences between treatment combinations. In instances where no Pb was detected in the leachate, no data bars are shown for that particular combination of treatment variables. Substrate codes are shown in the legend for Fig. 3

systems should also be closely examined as these soil conditioners may also introduce undesirable elements that may influence urban water quality.

Leaching of elements from green roof substrates

None of the substrates tested were significant sources of Cr, Cu, Fe, Ni, or Zn. A comparison of the leachate data here to the literature revealed that concentrations that leached from the substrates tested were comparable to reported values for various roof types, including vegetated roofs (Berndtsson et al. 2009; Berndtsson et al. 2006; Chang et al. 2004; Gnecco et al. 2005; Göbel et al. 2007; Mason et al. 1999; Zobrist et al. 2000). The leachate concentrations overall either did not exceed USEPA water quality standards or exceeded those standards no more often than other roof types. In fact, compared to some conventional roof materials, the substrates here were superior in that the percentage of samples that leached concentrations of Cu or Zn that exceeded the USEPA water quality standards was far lower than reported elsewhere (Chang et al. 2004) and nearly all of those samples that exceeded the standard were restricted to the first leaching (Figs. 6B, 8). Lassenite was the only substrate that could be considered a source of Mn. However one important detail of note is that the USEPA Recommended Water Quality Criteria for Mn does not reflect a value associated with Mn toxicity, but is instead a nuisance value that indicates the concentration at which undesirable flavors may appear in drinking water, or laundry stains may appear (USEPA 1999). Freshwater Mn concentrations up to

Table 2 Comparison of the maximum metal concentration observed in the leachate from pots across all treatments and leaching events in this study to metal concentrations for select conventional and vegetated roofs and to USEPA standards for water quality. The data from the literature are mean values for the measured dissolved metals for the indicated conventional roof type. In publications where more than one mean value was reported for a single roof type, the largest of the reported mean values was used. The standards represent either the USEPA freshwater quality criteria for priority (Cd, Cu, Ni, Pb, or Zn) or non-priority (Fe) contaminants, or the USEPA drinking water criterion for Mn. The percentage of samples from the planted and unplanted treatments, or from all treatments combined, that had leachate concentrations that exceeded USEPA water quality standards are indicated

	Cd	Cu	Fe	Mn	Ni	Pb	Zn
Maximum metal concentration, μg L^{-1}							
Leachate from pots	8.2	80.9	561.8	1,734	101.1	289.8	356.6
Mean metal concentration in roof runoff, μg L^{-1}							
Roof runoff							
Aluminum [a]	–	26	–	15	–	37	3,230
Copper [b]	0.8	2,600	–	–	4	69	370
Galvanized iron [a]	–	28	–	17	–	49	11,788
Gravel [c]	0.11	18	90	–	–	3	9
Gravel+plastic [d]	0.48	14.2	–	–	–	2.7	468
Polyester [c]	0.3	842	360	–	–	24	115
Slate+zinc gutters [e]	–	10	–	–	–	5.1	446.7
Shingle, wood [a]		29		44	–	45	16,317
Shingle, composition [a]	–	25	–	28	–	38	1,372
Tile [c]	0.4	304	415	–	–	41	48
Tile+concete [b]	0.8	153	–	–	4	69	1,871
Zinc [b]	1	153	–	–	4	69	6,000
Vegetated roof runoff [f]	–	600	5	–	–	2	10
Vegetated roof runoff [g]	–	88	60	62	–	2	1,209
Vegetated roof runoff [b]	0.1	58	–	–	3	6	468
USEPA Recommended Water Quality Criteria, μg L^{-1}							
Standards [h]	2	13	1,000	50	470	65	120
% of leachate samples from this study exceeding Water Quality Criteria							
- Plants	30.4	6.0	0	24.4	0	60.3	5.4
+ Plants	49.1	5.4	0	16.2	0	29.3	3.6
All pots	39.7	5.7	0	20.3	0	45.4	4.5

[a] Chang et al. 2004

[b] Göbel et al. 2007, data was taken from a review of studies of roofs with tile, concrete, fiber cement, bitumen, and glass or from a review of studies of intensive and extensive vegetated roofs

[c] Zobrist et al. 2000

[d] Mason et al. 1999, data obtained from analysis of combined runoff from a gravel roof, a gravel roof with a humus layer, and a plastic roof

[e] Gnecco et al. 2005

[f] Berndtsson et al. 2006, values estimated from the figures for an extensive vegetated roof in Sweden

[g] Berndtsson et al. 2009, runoff from an extensive vegetated roof in Sweden

[h] USEPA 1999

1,000 μg L^{-1} are not considered to be problematic in terms of toxicity to aquatic life (USEPA 1986). Only four leachate samples from the Lassenite pots exceeded this value.

The leaching of Cd and Pb from these substrates suggests that these materials could be potential sources of these heavy metals as there were a number of leachate samples that had concentrations of one or both of these elements in at least one leaching event that exceeded the relevant USEPA standards for water quality (Table 2). For unplanted systems, no single substrate produced leachate that consistently fell below the standards for both elements but Arkalyte, Haydite, Lassenite, and lava rock could be considered the best for Cd as these showed a first flush of that element and then minimal leaching for the second and third events. Complicating the situation was the observation that for most unplanted substrates, the leaching of Pb showed the opposite trend for these substrates with greater leaching in the later leaching events. The substrates that leached Pb were the same substrates that showed the highest total acid extractable and exchangeable Pb concentration, but the significant leaching of Pb from Axis and Haydite was unexpected given their lower total acid extractable and exchangeable Pb concentration (Figs. 1, 2).

These results underscore the need for a clear understanding of the geochemistry of metals, particularly USEPA priority contaminants, in each substrate and the time-dependence of their release. The decrease in leaching with time for some substrates may simply have been due to the depletion of a readily water-soluble pool or may represent a more complex response to the wetting-drying cycles that these substrates would experience within a green roof system. Repeated wetting-drying cycles are known to stabilize heavy metals (e.g., Cu, Cr, Ni, and Zn) within soils, shifting them from more labile to more stable pools with time (Han et al. 2001). Subjecting substrates to a series of wetting-drying pretreatments could in fact be beneficial in reducing the concentrations of elements that might readily leach from the substrate. Nevertheless, the behavior of Pb during the leaching experiment illustrates the potential importance of time and weathering to the release of heavy metals from green roof substrates. In soils, Pb demonstrates a limited solubility as it is typically associated with low solubility sulfides, clays, organic matter, and iron, aluminum, and manganese oxides (Riffaldi et al. 1976). Solubilization of Pb from these phases would require neutral to alkaline reducing conditions (Kabata-Pendias 2001). This element can also be found in phosphate, carbonate, or hydroxides precipitates that may be slowly soluble depending upon the local pH, or associated with slowly exchangeable sites on surfaces. In both of these cases, there would be a tendency for solubility to increase with time, perhaps as a function of the increase in the leachate volume across leaching events.

The release of inorganic elements from the green roof substrates could be further enhanced by the volume of water flowing through the green roof system and the chemical characteristics of the rainfall or stormwater that passes through the green roof systems and the volume of rainfall. The leaching study here used limited volumes of deionized water to provide a simple baseline for the leaching characteristics of these substrates. The volume of water used was not intended to mimic natural rainfall patterns but simply to prompt the leaching of water from the pots. Note from the results in Figs. 7 and 9 and the USEPA standards listed in Table 2 that mean values for leachate Cd and Pb exceed those standards by ~4-fold or less. Higher volume leachings, or more frequent leaching events, could readily reduce the concentrations of these elements in the leachate below the indicated standards, provided that the rainwater or stormwater did not further increase metal solubilization. In the urban environment, rainfall or stormwater could have dissolved solutes and organic carbon. The pH would also be expected to be slightly acidic as the average pH of uncontaminated rainwater is ≤5.6 (Charlson and Rodhe 1982). Increased acidity of urban rainfall or stormwater, or deposition of acid rain on green roofs, would perhaps be the most problematic scenarios as the solubility of the elements examined here generally increases as the pH decreases, particularly if the redox conditions are reducing rather than oxidizing (Kabata-Pendias 2001). Primary pollutants such as Cd, Pb, and Zn display this pH-redox relationship (Chuan et al. 1996). The mobilization of Cu and Zn from soils is also accentuated by dissolved organic carbon and pH (Kalbitz and Wennrich 1998; Sauvé et al. 2000; Zhao et al. 2007). Since there is limited information on the chemical composition of these substrates and distribution of metals (particularly Pb) within the matrices of those substances, the prospect for metal mobilization should be taken into account when considering these substrates for use in a green roof system.

The influence of plants on the leaching of metals from green roof substrates

An additional objective of this study was to determine whether the presence of a *Sedum* plant influenced the leaching characteristics of elements from these substrates. Here, the presence of a plant increased the leaching of Cd and Pb from Lassenite and the leaching of Cd from lava rock, bottom ash, and Haydite (Figs. 7, 9). The presence of a plant also produced interactions with the other main effects (i.e., substrate type and/or leaching event) to influence the leaching of Cu and Fe from some substrates (Figs. 4, 8), although not to an extent that significantly altered water quality with respect to these latter two elements. The only clear evidence that the presence of a plant significantly reduced leaching of metals was the results for Pb showing that in pots where a *Sedum* plant was present, Axis and bottom ash had a first flush of Pb, but negligible leaching thereafter and the results in Table 2 showing that only 29.3% of plant pots exceeded the USEPA water quality standard for Pb while the value was 60.3% for unplanted pots.

The lack of a consistent, positive contribution to water quality contrasts with studies that have reported benefits of vegetated roofs with *Sedum* plants with respect to the water quality of runoff (Berndtsson et al. 2009; Berndtsson et al. 2006; Köhler et al. 2002; Steusloff 1998). These studies differ from the context here in that the reported decreases in the metal concentration in the runoff were relative to the concentrations detected in rainfall. As noted in these studies, water retention is a contributing factor to the reduction provided by green roofs. The approach used here specifically encouraged leaching rather than collecting natural runoff or through fall, so may not have provided sufficient retention time for metal leaching to be ameliorated by the plants.

The possibility also exists that plants in green roof systems may enhance metal leaching by modifying the rhizosphere. Plant roots continuously release protons, organic acids, and chelating agents to facilitate nutrient acquisition (Marschner 1995). If micronutrient solubility from green roof substrates is low, or if these substrates are generally poor sources, then plants capable of modifying the rhizosphere will more proactively do so, with collateral effects on the solubility of inorganic pollutants (e.g., Fig. 7). This creates a potential conundrum in terms of water quality for those managing green roof systems. Micronutrient fertilization may be needed to minimize the extent to which plants in green roofs solubilize undesirable elements from the substrates, yet the fertilizers themselves may degrade water quality if they leach from the green roofs. However, it has been noted that conventional fertilizers may result in high nutrient concentration in green roof runoff water (Forrester 2007). Excess use of any type of fertilizers in a green roof project can result in runoff contamination (Berndtsson et al. 2009; Retzlaff et al. 2008). Phosphorus, for example, has been shown to leach from some fertilized green roof systems (Berndtsson et al. 2009; Berndtsson et al. 2006). Additional study is clearly needed to understand both the positive and negative contributions of plants to water quality in green roof systems.

Conclusions

Of the substrates examined in this study, none would be considered to be sources of Cr, Cu, Fe, Ni, or Zn in green roof systems as the concentrations detected in the leachate from the pots rarely (<8% of samples across all treatment combinations) exceeded USEPA standards for water quality. All substrates except Lassenite met the USEPA water quality criteria for Mn (50 μg L^{-1}) and therefore are not sources of Mn. However, this particular standard may not provide a valid basis comparison as it is not based upon Mn toxicity. When leachate Mn concentrations from Lassenite were compared to a toxicological standard for Mn, namely 1,000 μg L^{-1} for toxicity to aquatic organisms (USEPA 1986), only four of the samples collected exceeded that standard. If that standard is considered, Lassenite would also not be a source of Mn. Nevertheless, the tendency of this substrate to leach Mn deserves additional scrutiny. All of the substrates tested produced leachate with Cd and/or Pb concentrations that exceeded the USEPA water quality standards during at least one sampling event during the leaching study and are potential sources of these metals. The elevated concentrations of Pb in several of these substrates clearly contributed to these results, but additional biogeochemical processes not considered here may have influenced metal leaching. The limited volume of water used to leach the pots may also have resulted in leachate metal concentrations that were higher than might be observed in established green roof systems exposed to more frequent, higher volume precipitation events. Increased flow volume might be all that is needed to dilute the metals in the leachate from some of the substrates tested to concentrations below the applicable USEPA standards. Leaching and water retention studies with these substrates will be needed to determine if metal leaching is related to retention, as has been observed in previous studies (Berndtsson et al. 2006; Steusloff 1998). Whether increased flow through these substrates will provide the necessary dilution or exacerbate the leaching of Cd and Pb must be determined.

Batch to batch variation in these substrates must also be addressed. The starting materials for these substrates are heterogeneous and the elemental content would be dictated by the mineralogy at the location from which the raw material was mined. If production or processing methods for these substrates are not standardized, or if there are no quality standards that needed to be met for a substrate, then a definitive statement about whether these materials would be metal sources in green roof systems cannot be made without additional study. At a minimum, batches of substrates with lower exchangeable metal concentrations should be selected to provide the most direct means of limiting the release of metal pollutants. If substrates can be identified that are not sources of metal pollutants, the next step would be to identify those capable of adsorbing and/or sequestering pollutants introduced by rainfall, or at least restricting their release into runoff.

The presence of a single *S. hybridum* plants in the planted treatments increased leaching of Cd, Pb, and other metals from some substrates yet greatly reduced Pb leaching from other substrates. Additional, longer term experiments are required to examine more comprehensively the impact of plant roots and root exudates on the solubilization of inorganic pollutants from green roof substrates and the ability of plants to sequester soluble metals released from substrates used in vegetated roofs. The degree to which green roof systems serve as sources and sinks of pollutants is clearly an important aspect of their contribution to the health of the urban environment. If plants can be identified that restrict rather than promote pollutant release, and these substrates are coupled with substrates that promote plant growth while also reducing pollutant leaching, then green roofs systems capable of improving urban water quality can be developed.

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